AIP Journal of Applied Physics

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Citation: Journal of Applied Physics **66**, 47 (1989); doi: 10.1063/1.343854
View online: http://dx.doi.org/10.1063/1.343854
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/66/1?ver=pdfcov
Published by the AIP Publishing

Phase-shift measurement in photorefractive holographic recording

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(Received 26 October 1988; accepted for publication 12 April 1989)

A simple direct method for measuring the interference pattern of a light-to-recorded hologram phase shift in photorefractive crystals is reported. The method is used for studying the recording in $Bi_{12}SiO_{20}$ under an externally applied electric field. The results are then compared with theory and used for computing the density of photoelectron traps.

I. INTRODUCTION

The interference pattern of light projected onto a photorefractive crystal and the resulting refractive-index modulation volume hologram are, in general, phase shifted.[1] The amount of such shifts depends on the nature of the recording mechanism involved: A purely diffusion-controlled recording results in a $\pi/2$ shift, while a predominantly drift-controlled recording under the action of an externally applied electric field may considerably depart from that value. The knowledge of such phase shift may therefore provide one with interesting information concerning the building up and the nature of the space-charge arising electric field during holographic recording under an externally applied field. The practical interest of this phase shift is due to the fact that it does control energy transfer in beam coupling experiments: Optimal transfer occurs for $\pi/2$, while there is no transfer at all for 0 and π phase shifts.[2]

Energy exchange, however, is not only limited by phase shift but also depends on diffraction efficiency and on rather complicated optical polarization relations between the diffracted and transmitted beams through the crystal.[3] Recording under an applied electric field results in a higher diffraction efficiency,[4] but an unwanted departure from optimal $\pi/2$ conditions results as well. Anisotropic diffraction properties[5] characteristic of photorefractive recording, optical activity of most crystals, and the bulk anisotropy produced by the applied electric field result in a complicated polarization dependence of energy transfer in terms of the applied field. That is why the phase-shift dependence on the applied field is not usually available simply from energy-exchange experiments.

In this paper, however, we show that it is possible to compute the interference pattern-to-hologram phase shift in photorefractive crystals from simple two-wave mixing experiments, regardless of wave polarization states, diffraction efficiency, laser beam intensity variations, and photodetector system characteristics. Thus we measured the phase-shift dependence on the applied electric field in a $Bi_{12}SiO_{20}$ sample. We then show that experimental results agree with theory and use these data for computing the density of photoelectron traps.

II. INTERFERENCE TERM IN TWO-WAVE MIXING EXPERIMENTS

In a recent paper,[6] we showed that phase modulation of one of the interfering beams in a two-wave mixing experiment (Fig. 1) allows synchronous detection of the first (I_Ω) and the second ($I_{2\Omega}$) harmonic terms of the resulting output irradiance I_R (or I_S) through the photorefractive crystal. Their mathematical formulation is

$$I_\Omega = 2\psi_d\sqrt{I_1 I_2}\sqrt{\eta}\, F\cos\psi \tag{1}$$

and

$$I_{2\Omega} = 2(\psi_d/2)^2\sqrt{I_1 I_2}\sqrt{\eta}F\sin\psi, \tag{2}$$

where I_1 and I_2 are the irradiances of each interfering beam, η is the diffraction efficiency of the hologram (assumed to be $\eta \ll 1$) in the crystal, ψ_d is the amplitude of the phase modulation at frequency Ω, F is the wave-polarization electric field-dependent term which assumes a simple form only for particular experimental conditions,[6] and ψ is the phase shift we want to measure. Both terms I_Ω and $I_{2\Omega}$ are measured using the same detector but with two independent lock-in amplifiers tuned to Ω and 2Ω, respectively. Dividing I_Ω by $I_{2\Omega}$, substituting ψ by $\pi/2 + \phi$, and rearranging terms, we get

$$\tan\phi = (I_\Omega/I_{2\Omega})(\psi_d/4). \tag{3}$$

As deduced from Eqs. (1) and (2), the $I_\Omega/I_{2\Omega}$ ratio is not at all dependent upon either I_1 and I_2, η, or the polarization term F. The polarizer in the setup (Fig. 1) is intended only for choosing the incident polarization in order to have the diffracted and transmitted beams at the crystal output parallel polarized for no external field,[6] which increases the signal and eases measurement. No particular requirements for the photodetector in the experiment are needed except for a reasonable flat response in the Ω–2Ω frequency range.

III. THEORETICAL PHASE-SHIFT DEPENDENCE ON APPLIED EXTERNAL FIELD

Assume a holographically generated interference pattern of light of period $\Delta = 2\pi/K$,

$$I = I_0(1 + m\cos Kx), \tag{4}$$

projected onto the (110) face of a $Bi_{12}SiO_{20}$ crystal in a transverse electro-optic configuration with the x axis along the $[1\bar{1}0]$ crystal axis and parallel to the grating vector $\mathbf{K}$. The resulting steady-state first-harmonic-term space-charge arising electric-field modulation amplitude is found to be[6,7]

$$E_{sc} = -m\frac{E + iE_D}{1 + E_D/E_q - iE/E_q}, \tag{5}$$

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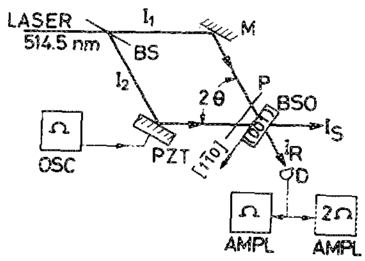

FIG. 1. Experimental setup. An oscillator (OSC) produces a low-amplitude phase modulation at frequency $\Omega/2\pi = 1500$ Hz on the holographic pattern through the piezoelectric supported mirror (PZT). BS is the beam-splitter, and AMPL are lock-in amplifiers tuned to Ω and 2Ω.

where E is the externally applied field, $E_D \equiv K(k_B T/q)$ is the purely diffusion space-charge arising electric-field amplitude in the crystal, $E_q \equiv qN_A/(K\epsilon)$ is the maximum amplitude of the electric-field modulation assumed to be purely sinusoidal and limited only by the electron trap density N_A, k_B is the Boltzmann constant, T is the absolute temperature, q is the electron charge, ϵ is the electric permittivity, and m is the interference fringe contrast. From Eq. (5) and from $\tan\phi = \tan(\psi - \pi/2) = -1/\tan\psi$, we get

$$\tan\phi = \frac{EE_q}{E^2 + E_D E_q(1 + E_D/E_q}). \tag{6}$$

The highest value for ϕ may be obtained from Eq. (6) simply by making $\partial \tan\phi/\partial E = 0$, resulting in

$$(\tan\phi)_M = \frac{1}{\sqrt{1 + E_D/E_q}} \frac{q}{2K} \sqrt{N_A/(\epsilon k_B T)} \tag{7}$$

for

$$E_M^2 = E_q E_D\left(1 + \frac{E_D}{E_q}\right) = \left(1 + \frac{E_D}{E_q}\right) N_A \frac{k_B T}{\epsilon}. \tag{8}$$

Substituting Eqs. (7) and (8) into Eq. (6), the latter may be now written as

$$\tan\phi = [2(\tan\phi)_M E_M/(E_M^2 + E^2)]E. \tag{9}$$

The parameter E_q may be computed from Eqs. (7) and (8):

$$E_q = qN_A/(\epsilon K) = 2(\tan\phi)_M E_M, \tag{10}$$

and the associated trap density N_A may be accordingly obtained knowing that[8] $\epsilon/\epsilon_0 \sim 56$ for BSO.

IV. EXPERIMENTAL RESULTS

We measured $\tan\phi$ against the applied field E for $\theta = 10°$ ($K = 4.24\ \mu\text{m}^{-1}$), 20° ($8.35\ \mu\text{m}^{-1}$), and 24° 20′ ($10.06\ \mu\text{m}^{-1}$), as proposed in Sec. II (see Figs. 2 and 3). Experimental data were fitted to Eq. (9), and the resulting $(\tan\phi)_M$ and E_M were thus obtained. The corresponding parameters E_q, N_A, and E_D were computed as referred to above. All such data are reported in Table I.

Note that the self-diffraction technique used in our experiment and Bragg limitations characteristic of these volume holograms lead us to measure just the first-harmonic term referred to in theory and represented by Eq. (5). The influence of higher harmonic terms upon the first one, however (not accounted for in the current theoretical approach), can actually be neglected using either low-contrast interference holographic fringes ($m \ll 1$) in the experiment, or working far from saturation conditions [that is, $E \ll E_q$, which substituted into Eq. (5) with $m \simeq 1$ and $E \gg E_D$ results in a far-from-saturation space-charge electric-field modulation $E_{sc} \simeq E \ll E_q$]] in order for the linear superposition principle to hold for all harmonic terms actually present. Accordingly, experimental data are expected to fit theory better for $m \simeq 0.3$ (unfortunately, lower values for m represent lower signal-to-noise ratio (SNR) as well, which strongly complicates experimental measurement) than for $m \simeq 1$. In fact, distinctly higher (and presumably more reliable) values for N_A are reported for $m \simeq 0.3$ in Table I. Better agreement with theory should have also been expected for large E_q ($E_q \propto 1/K$) experimental conditions, a fact that is not apparently supported from the experimental results in Table I, at least in the small $\theta = 10°$ to $\theta = 24°\ 20'$ range reported here.

It is also worth noting that a constant abscissa shift (~ 250–~ 900 V/cm as reported in Table I) is needed for better data fitting. Otherwise sensible differences in computed values for E_D and E_q are obtained. We think this shift may be due to the imperfect electric contact characteristic of the colloidal silver electrodes used in the experiment. We have no evidence that this shift is due to any weak photovoltaic effect instead, because no sensible phase shift was ever detected for $E = 0$ in this work. This matter, however, deserves further research.

V. CONCLUSIONS

For the first time, to our knowledge, a simple and reliable method for measuring ϕ is proposed. The method is used for measuring ϕ as a function of the applied electric field for a $Bi_{12}SiO_{20}$ sample in a two-wave mixing experiment. The method is not sensitive to diffraction efficiency

TABLE I. Experimental results.

		E_D (kV/cm)		E_q	N_A	E_M		Abscissa shift
θ	m	Theor.	Exp.	(kV/cm)	(10^{22} m^{-3})	(kV/cm)	$(\tan)_M$	(kV/cm)
10°	1	1.80	1.70	11.51	1.50	4.74	1.21	0.32
20°	1	2.13	2.70	10.36	2.65	5.94	0.87	0.26
24° 20′	1	2.57	2.25	7.64	2.35	4.72	0.81	0.92
10°	0.3	1.08	1.47	30.50	3.96	6.86	2.22	0.23
24° 20′	0.3	2.57	2.51	9.26	2.85	5.44	0.85	0.65

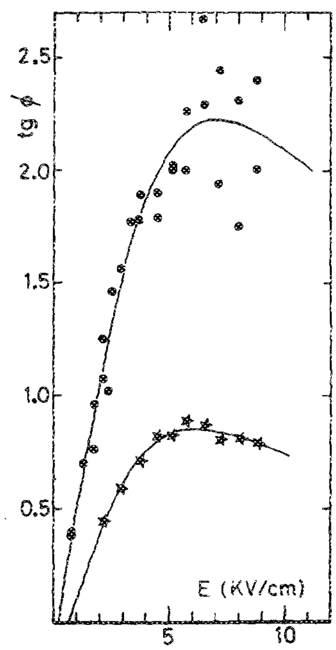

FIG. 2. Experimental tan ϕ vs E data for $K=4.24\ \mu m^{-1}$ (⊗) and $10.6\ \mu m^{-1}$ (✵), always for $m \simeq 0.3$, with $I_1 \simeq I_2/50$ and $I_2 \simeq 600\ \mu W/cm^2$. Continuous lines are the best fitting to theory.

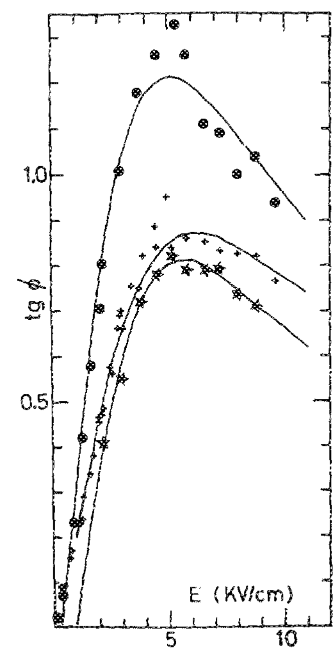

FIG. 3. Experimental tan ϕ vs E data for grating vectors $K=4.24\ \mu m^{-1}$ (⊗), 8.35 μm^{-1} (+), and 10.06 μm^{-1} (✵), always for $m \simeq 1$ and roughly $I_1 \simeq I_2 \simeq 600\ \mu W/cm^2$. Continuous lines are the best fitting to theory.

variations (including those arising from instabilities related to the "running hologram"[7] phenomenon characteristic of holographic recording under applied electric field), beam intensity changes, and wave polarization relations, which seriously limit other methods.

The successful matching of experimental points to the theoretical curves supports this method and, in adequate experimental conditions, allows one to compute the relevant photoelectron trap density in the crystal. Note, however, that a good agreement of experimental and theoretical E_D values was obtained only for one of the five cases reported in Table I. It should also be noted that, in spite of the general assumption that a reasonable good agreement with theory may be obtained from $m \simeq 0.3$ experiments, sensible differences in the computed N_A values were actually obtained in such conditions for $\theta = 10°$ and $\theta = 24° 20'$ (Table I). The average $N_A = 3.4 \times 10^{22}\ m^{-3}$ value for $m \simeq 0.3$ is roughly half the value we have already reported ourselves for the same sample using a holographic erasure technique.[9] Such a difference may arise from the fact that the positively ionized shallow electron trap centers that we are actually measuring here may be assumed to have a constant density N_A but only for low irradiance level experiments (which is neither the case here nor in Ref. 9). Otherwise some variation may be expected particularly for the roughly 10–30-fold higher irradiances in Ref. 9 as compared to those in this work.

Most interesting in this method, however, is the possibility of measuring ϕ even in conditions where the current theoretical approach to photorefractive crystals phenomena may not verify at all.

ACKNOWLEDGMENTS

We acknowledge S. I. Stepanov, A. A. Kamshilin, Elena Makrushina, and V. V. Kulikov from the A. F. IOFFE Physico-Technical Institute in Leningrad, USSR, for useful discussions. This work was supported by Financiadora de Estudos e Projetos, Conselho Nacional de Desenvolvimento Cientifico e Tecnologico, and Coordenadoria de Aperfeio coamento do Pessoal de Ensino Superior.

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